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PROPERTIES OF ORGANIC DRIVING MEDIA FOR DIFFUSION PUMPS IN TECHNICAL OPERATION AND A NEW FORM OF AN OIL DIFFUSION PUMP

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PROPERTIES OF ORGANIC DRIVING MEDIA FOR DIFFUSION PUMPS IN TECHNNICAL OPERATION AND A NEW FORM OF AN OIL DIFFUSION PUMP

R. Jaeckel[1]

ABSTRACT. Advantages and disadvantages of oil diffusion pumps over mercury diffusion pumps are discussed in relation to the use of different types of organic driving media, These include apiezonoils, diesters of phthalic acid and liphatic dibasic acids. Methods of use, operating properties and results are discussed.

Diffusion pumps driven by organic media (so-called diffusion pumps) have /177* appreciable advantages over mercury diffusion pumps. The danger of poisoning the people working with the pumps is totally avoided. The inclusion of a freezing device between the pump and the instrumentation and the resulting throttling of the suction velocity is eliminated. In certain situations, for instance in the distillation of materials designed for human or animal consumption, there is another advantage in that in using mercury the actual danger of contaminating products may occur. In conclusion it is also preferable in light of making our domestic economy independent to use oil diffusion pumps employing domestic oils rather than the rarer metal mercury. On the other hand, there are some obvious advantages. The oil diffusion pumps require a higher fore-vacuum than mercury diffusion pumps. But in the case of the high vacuum values obtained by means of modern forepumps (rotating oil air pumps) this is not a serious problem any longer. A disadvantage of the oil diffusion pumps is the fact that the organic driving media (oils) are not as resistant (for instance, to oxidation resulting from sudden air venting into the instrumentation) as mercury. The sensitivity of the oils is very often exaggerated, however. This will be proven subsequently on the basis of experiences with oil /178 diffusion pumps in actual operations. Moreover a comparison will be drawn between the properties of different organic driving media and in conclusion

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[1]From the high-vacuum laboratory of E. Leybold's Successors Company, Cologne-Bayental.

*Numbers in the margin indicate pagination in the foreign text.

we shall describe a form of oil diffusion pump which can substantially overcome the difficulties resulting from the instability of the oils.

I. Type of Driving Media and Measurement Method for the Determination of the End Vacuum

a) Type of Driving Media

Two groups of materials assume a particular technical significance in the evaluation of organic driving media for diffusion pumps.

I. Apiezonoils

Apiezonoils [1] are mineral oil fractions of the lowest possible vapor pressure obtained by the short path distillation in high vacuum. Until 1939 they were manufactured in England by the Asiatic Petroleum Company, and by the firm Leybold's Successors, Cologne in two types: Apiezonoil A and Apiezonoil B has the lowest vapor pressure of both types with a value of 10^{-7} Torr at room temperature. Since 1940 the Leybold Company has been manufacturing products equivalent to the Apiezonoils and sells them under the name of Leybold--Apiezonoils D and E.

II. Diesters of the Phthalic Acid [2] or of the Aliphatic Dibasic Acids [3], Particularly of Sebacic Acid, with Higher Alcohols

Similar uniform organic materials[2] were first used by Hickmann [2] as driving media for diffusion pumps. Distillation Products, Inc., Rochester, New York, in the United States, a subsidiary of Kodak, markets a phthalic acid ester, the 2-ethylhexylphthalate under the trade name Octoil and the corresponding ester of the 2-ethylhexylalcohol with sebacic acid as Octoil S. Octoil and Octoil S have a room temperature vapor pressure of approximately 10^{-7} Torr[3] and the same value applies also for Apiezonoil B and Leybold-Apiezonoil E.

We could assume that the reachable end vacuum of an oil diffusion pump system would be solely determined by the vapor pressure of the driving media at the cooling temperature. Consequently we should be able to obtain an end

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[2]The Apiezonoils are chemically not a uniform material but a mixture containing a large proportion of saturated hydrocarbons.

[3]Other indicated values originated from measurements (see below).

vacuum of 10^{-7} Torr with any oil diffusion pump system operating with Apiezonoil B, Leybold-Apiezonoil E or Octoil. But this is not the case.

Before we enter into the reason for this, we should subject the measurement methods under which the following investigations for the determination of end vacuum have been conducted to a brief review.

b) Measurement Methods for the End Vacuum and Determination

There are fundamentally two types of vacuum gauges available for the end vacuum measurement in the pressure range from 10^{-3} to 10^{-7} Torr, namely the ionization manometer and the absolute manometer (Knudsen).

I. Ionization Manometer

Practically all end vacuum measurements on all diffusion pumps described in the literature have been performed with ionization manometers. In these vacuum gauges, as we know, we do not measure the pressure (p), directly, but the relationship of the electron flow ($J^=$) to the ion flow (J^+) generated by the pulse ionization of the electrons.

$$p = C \frac{J^+}{J^-} \qquad (1)$$

The factor C depends on:

1. The geometric configuration of the tube where the measurement is effected and,
2. The effective cross-section against the ionizing electron pulse of the gas or vapor molecules present in the tube space (that is, the gas type).

The dependence of the geometric configuration results from the fact that the effective cross-section in the case of different molecules will depend in different ways on the electron velocity at the point of impact.

The end vacuum measurement could also be effected, as it has been many times observed, where the factor C may be determined, for instance, by calibration with air and then the end vacuum determination would be made with the help of the value of p determined from equation (1) and multiplied with

the relationship

$$\frac{\text{Effective Cross Section in Air}}{\text{Effective Cross Section in Vapor}} .$$

But, since the effective cross-sections could best be estimated[4], we intentionally deviate from these apparently rational methods and in their stead indicate for all end vacuum measurements the value of p, as it results from equation (1) according to the relationship of the currents J^+/J^- and the constant C for air. The measurements were effected with a Telefunken tube (RE 11). The calibration with air yielded a value of C = ±0.28 [4]. /179

II. Molecular Vacuum Gauge

Although the end vacuum determinations with ionization manometers according to the above statements should appear problematic, we have used ionization manometer measurements throughout for comparison with the values described in the appropriate literature[5].

The absolute manometer delivers an immediate pressure indication when reaching the end vacuum (according to Knudsen) [5]. In this instrument the molecules emitted by a hot wall exercise a pressure on one side of a moveable membrane which is counteracted by molecules reaching the other side and obviously depends (if we exclude accommodation coefficients) only on the number of molecules per unit volume of the gas space (n) and not in the type of gas, since

$$p = \frac{nmc^2}{3}$$

and mc^2 is independent of the type of gas.

The absolute manometer [5] was used in the form of a molecular vacuum gauge [6] as described by Gaede.

[4]In the case of the ionization manometer measurement there is an additional difficulty resulting from the fact that upon an electron pulse and depending upon the type of molecules present in the gas space, the formation of the least energetic of the combinations of a molarion and one electron, or of a neutral molecule, a molarion and one electron or also of two molarions is possible.

[5]Such a comparison has only a limited value because of the above mentioned influence of the geometrical configuration of the measuring tube, since the different authors have in each instance worked with different types of measuring tubes.

2. End Vacuum

(Value obtained in the most favorable case with a specific pump system of attainable value).

As we have already indicated in the introduction, the end vacuum obtainable with a given driving medium is not only dependent upon the type of the driving media (that is, its vapor pressure). The achievable end vacuum is instead initially dependent on the pumping installation, whether individual pumps or pump banks, metal diffusion or glass diffusion pumps are used as well as the operating conditions, fore-vacuum and temperature in the distillation flask (that is, the heat energy of the electrical heating element).

The dependence of the end vacuum on the particular type of pump in which the driving medium is used is shown in Table I. According to this table, for instance, Apiezonoil B shows the worst end vacuum in a single metal diffusion pump, a better one in a pump bank of two back-to-back metal diffusion pumps and the best in a series of glass diffusion pumps.[6]

This behavior is easy to understand when one considers that Apiezonoil is not a chemically uniform substance but a mixture of different products with different molecular weights and boiling points. In a pump series, however, the components with the highest vapor pressure will distill gradually from the high vacuum stage into the stage functioning as forepump. The high vacuum stage will then contain only the components with the lowest vapor pressures and consequently the value of the end vacuum will improve. /180 There are two more phenomena. All organic driving media are neither completely resistant to thermal decomposition nor to oxidation. The products formed by both influences have in part higher vapor pressures than the original driving medium. These products also distill in the forepump of a pump bank. The oxidation sensitivity of the driving media also further explains the fact that in general the end vacuum obtained with oil diffusion pumps improves as the pressure on the forepump side is reduced. This also reveals the superiority of a pump series in opposition to an individual pump, since in a pump series the fore-vacuum of the high vacuum stage results from a diffusion pump rather than from a rotating

[6]A pump series or bank is an arrangement of several back-to-back diffusion pumps.

oil-air pump. This will give much lower values than in the case of an individual pump. Finally, the thermal decomposition is favored by the presence of metals. This explains why glass pumps are able to reach better end vacuum values.

The factors introduced in the above example of Apiezonoil B as the critical factors for the end vacuum of an oil diffusion pump influence in similar fashion the end vacuum of Octoil. In the case of Octoil it appears that originally it was a chemically uniform substance. Yet, it still contains traces of impurities resulting from its manufacture, and the uniformity of the material is also constantly disturbed by the thermal decomposition and oxidation. The large diffusion pumps (see Table I, particularly the measurement with the ionization manometer) indicates considerable thermal decomposition in the case of Octoil. This agrees with the findings of Kodak for the end vacuum attainable on the one hand with Octoil in pure glass pumps and on the other hand in glass-metal pumps (see Table II). The difference between the absolute values of our measurements and the Kodak data results, on the one hand, from the fact that the measurements were not performed with exactly identical pump systems. In the first place, we worked with a glass-metal pump. In the second place our measurements were performed with a single pump series consisting of two back-to-back glass diffusion pumps, whereas the Kodak measurements were made with a three-stage glass diffusion pump of special design.[7] Moreover, we calculated our end vacuum values based on the ionization manometer measurements calibrated in air in accordance with equation (1), while Kodak apparently simply multiplied the ratio of the effective cross-section of air and Octoil [7].

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[7]Pump with separate boiling flasks (Hickmann design), (see below for further details.)

TABLE I. END VACUUM VALUES OBTAINED

Pump Installation	Apiezonoil B measured with		Octoil measured with	
	Ionization manometer Torr	Molecular vacuum gauge Torr	Ionization manometer Torr	Molecular vacuum gauge Torr
Metal diffusion pump*	$1.2 \cdot 10^{-5}$**	$2 \cdot 10^{-6}$**	$4 \cdot 10^{-5}$†	$7 \cdot 10^{-6}$†
Metal diffusion pump series	$4 \cdot 10^{-6}$			
Glass diffusion pump series††	$1.7 \cdot 10^{-6}$	$8 \cdot 10^{-7}$	$1.1 \cdot 10^{-6}$	$2.1 \cdot 10^{-6}$

*Two-stage oil diffusion pump (Leybold, Model P), Forevacuum pump (Leybold, Model V).
**Heat Energy 450 Watt.
†Heat Energy 250 Watt.
††Heat Energy 300 Watt. Forevacuum Leybold, Model V.

The measurements with the ionization manometer indicated higher pressure values than the measurements with the molecular vacuum gauge because in the case of the molecular vacuum gauge the pressure is measured directly while the ionization manometer measures only an ionation and the cross-section of the heavy vapor molecules against an electron pulse ionization which is naturally larger than the effective cross-section of the air molecules. Since both the ionization manometer and the molecular vacuum guage were calibrated with air, the ionization manometer measurement at constant oil vapor temperature should yield a higher pressure value than the molecular vacuum measurement.

The dependence of the end vacuum of a diffusion pump from the heating energy is shown in Figure 1. The thermal decomposition increases with increasing heating energy and the end vacuum pressure rises[8].

[8]The end vacuum values given below were determined in such a manner that firstly the optimal heating energy in accordance with Figure 1 was determined and then the end vacuum corresponding to that heating energy was indicated. Two-stage rotating oil air pumps were used in all instances as forevacuum pumps (Leybold-Models V and XII).

TABLE II. END VACUUM VALUES REACHED WITH OCTOIL
(Measured with the Ionization Manometer)

No.	Pump Installation	Kodak Measurements Torr	Pump Installation	Kodak Measurements Torr.
1	Glass-metal pump	$5 \cdot 10^{-6}$	Metal diffusion pump	$4 \cdot 10^{-5}$
2	3-stage glass diffusion pump with separate distillation flasks	$2.5 \cdot 10^{-7}$	Glass diffusion pump series	$1.1 \cdot 10^{-6}$

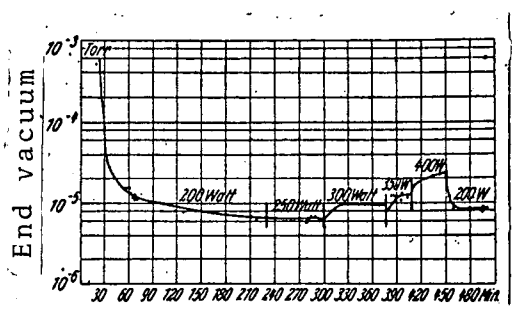

Figure 1. End Vacuum Pressures Obtained With a 2-Stage Metal Diffusion Pump (Leybold Model P) Driven by Octoil at Different Heating Energies. (End vacuum measured with the molecular vacuum gauge, forevacuum pump Leybold Model V).

The above-described superiority of the diffusion pump series in contrast to single pumps results, as it was suggested, from the self-cleaning of the oil by the distillation process in the pump installation. Hickmann [8] has described a multi-stage single pump which deliberately promotes these distillation processes by regulating the oil circulation in the pump. He has thus applied the principle first expressed by Gaede [9] of the self-cleaning of oils by deliberate encouragement of distillation processes. The Hickmann pump /181
has subsequently been modified often [10]. The principle of all these pumps is briefly as follows: multiple back-to-back jets installed between the forevacuum and the high vacuum receive their oil vapor from separate boiling flasks, which however, share a common liquid supply. The oil first flows as a liquid through the boiling flask of the stage immediately adjacent to the forevacuum. There

the lowest boiling fractions of the oil evaporate and serve to operate the first stage. The oil freed of these lighter fractions then flows into the liquid space of the boiling flask of the second stage. There the next higher boiling fractions of the oil evaporate and serve to operate the second stage, etc. Hence, the last stage, closest to the high vacuum, is reached only by the highest boiling component with the lowest vapor pressure.

This Hickmann arrangement does have a major disadvantage. All the oil vapor fractions, after they have flowed through the jets and condensed on the cooling surface, return to the boiling flask of the first stage and consequently to the vicinity of the forevacuum. According to the above conclusions on the influence of the forevacuum on the end vacuum, a severe threat of oxidation exists. To avoid these errors, pumps have been described in which the oil, after it flows through the jets in vapor form and subsequently condenses can be returned to the boiling flask of the same stage. This, however, has the disadvantage that the thermal decomposition products formed during the evaporation will also be introduced into the circulation. In accordance with the French patent 838111 an attempt has been made to prevent this by fractional condensation. A fractional condensation in high vacuum is quite impossible, however, because of the long free path lengths of the vapor molecules.

We have consequently developed a multi-stage oil diffusion pump with separate boiling flasks[9], which uses the Gaede principle of oil fractioning in the pump, but which avoids the above-described disadvantages of the Hickmann pump and its modifications. Figure 2 shows a schematic cross-section of such a pump. The high-vacuum chamber is connected at 21 and the forevacuum at 22. The three back-to-back pump stages 15, 16, and 17 receive their oil vapor from separate boiling chambers 5, 6, and 7. The oil condensing on cooling surface 23 arrives by opening 9 first in the boiling flask of the first stage. There it vaporizes the lowest boiling components of the oil which serve to activate stage 15. The oil freed of the lighter components now arrives through opening 10 in boiling flask 6 of the second stage. There the medium weight components of the oil vaporize. The generated vapor operates stage 16.

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[9]This pump is sold by the E. Leybold and Successors Company of Cologne-Bayental as oil diffusion pump Model Q3.

Finally the oil arrives through opening 11 in the boiling flask 7 of the high-vacuum stage 17 completely free of the light and medium weight fractions. Stage 17 operates with the highest boiling fractions of the oil, i.e., those with the lowest vapor pressure.

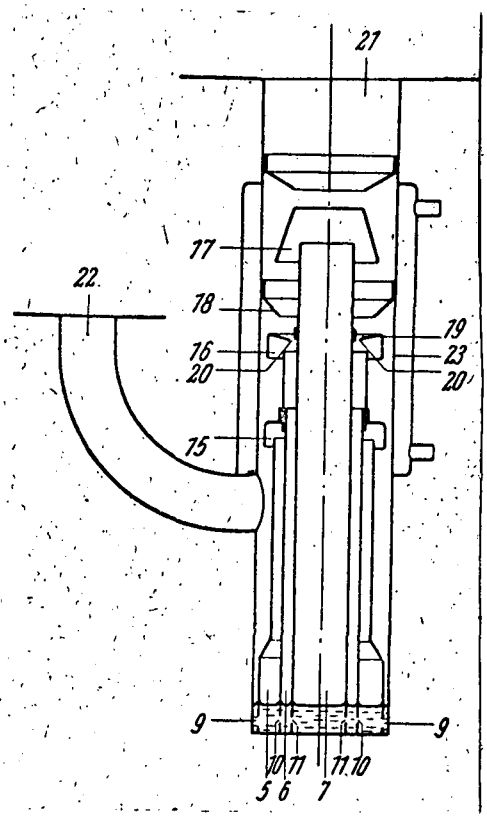

Figure 2. Cross-Section (Schematic) Through a 3-Stage Metal Oil Diffusion Pump With Separate Boiling Flasks and Oil Reflux to the Second Stage (Leybold Model Q3).

In contrast to the Hickmann pump these high-boiling, therefore most valuable, oil components do not return to the boiling flask 5 after flowing through the jets 17 and condensing on the cooling surface 23 but are diverted by the baffle plate 18 into the oil groove 19, then reaching the boiling flask 6 of the second stage through opening 20. The light and medium heavy components /182 formed by the thermal decomposition and oxidation vaporize there. Hence, the boiling flask 7 of the high-vacuum stage is reached only by the highest boiling components: externally this pump differs from the Hickmann arrangement only by the presence of the baffle plate 18, and oil groove 19 and the oil reflux opening 20. The major advantage is that in this manner it is possible to

prevent the reflux of valuable high boiling components to the boiling flask 5 of the first stage and consequently the inter-mixing of the light fractions and the increased danger of oxidation. The critical factor for the efficiency of the pump, is, moreover, that the reflux of the heavy oil components does not occur into boiling flask 7 of the high-vacuum stage proper but instead into the boiling flask 6 of the preceeding stage, so that in this way the light and medium weight fractions formed by chemical decomposition and oxidation are kept away from the high-vacuum stage.

Table 3 shows the end vacuum values achieveable with this pump. The operation of this pump with Apiezonoil consequently allows higher end vacuums with a simple metal diffusion pump. The end vacuum obtained has approximately the same value as that obtained by a metal diffusion pump series. But even the operation of the pump with ordinary machine oil permits the achievement of very useful end vacuum values of approximately 10 Torr. The operation with machine oil does however have the disadvantage that during the first breaking-in of a new oil filling appreciably long warmup times of approximately 30 hours (3) are necessary to achieve a vacuum of 10^{-5} Torr. By comparison, Figure 4 shows the warmup time of a single metal diffusion pump operating with Apiezonoil. The end vacuum values obtained with a pump with separate distillation flasks, as in Figure 2, are compared in Table III with the performance of a metal diffusion pump series. The suction velocity of the pump has approximately the same value of 20 liters/sec as the metal diffusion pump series used in the comparison. On theother hand, this pump requires for that performance a heat energy of only approximately 400 watts as opposed to the 750 watts required for the diffusion pump series. The high forevacuum required (0.1 Torr) to reach the end vacuum with this pump has, in spite of the much smaller heat requirement, approximately the same value as for the metal diffusion pump series (0.4 Torr). In contrast a singular metal diffusion pump of conventional construction at the suction velocity of 20 liters/sec and a heat requirement of 320 watts requires for the achievement of the end vacuum of a forevacuum better than 0.01 Torr. An added advantage of the pump of Figure 2 lies in the rapid increase of the end vacuum after venting; compare Figure 8 and Figure 7.

In addition to the technically important organic driving media for oil diffusion pumps Apiezonoil and Octoil the literature reveals a substantial number of other driving media, all of which yield less favorable results, however, Table IV contains the end vacuum reached with some of these driving media in the case of a single stage metal diffusion pump and a comparison with Apiezonoil.

TABLE III. END VACUUM VALUES
(Measured with the Ionization Manometer)

Pump Arrangement / Oil Filling	Apiezonoil B Torr	Machine Oil Torr
Metal diffusion pump	$1.2 \cdot 10^{-5}$	approx. 10^{-4}
Metal diffusion pump series	$4 \cdot 10^{-6}$	----
Pump with separate boiling flasks according to Figure 2.	$4 \cdots 6 \cdot 10^{-6}$	After 30 operating hours $1 \cdot 10^{-5}$ extreme limit $7 \cdot 10^{-6}$ *

*This value should be compared with that given by A. E. Lockenvitz, Rev. Sci. Instr. 8 (1937) 322, which was not reached with a 6-stage pump built in accordance with the Hickmann model.

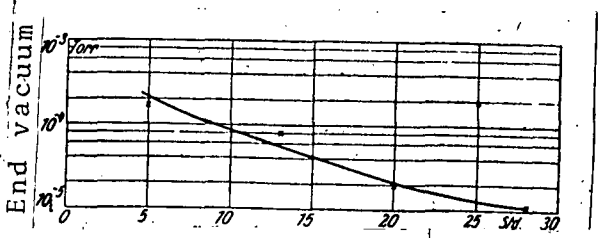

Figure 3. Three-stage Metal Oil Diffusion Pump with Separate Boiling Flasks and Oil Reflux to the Second Stage (Leybold Model Q3, Operating with Machine Oil (Shell CY2). (Heat Energy 410 W, end vacuum measured with an ionization manometer, forevacuum pump Leybold Model V).

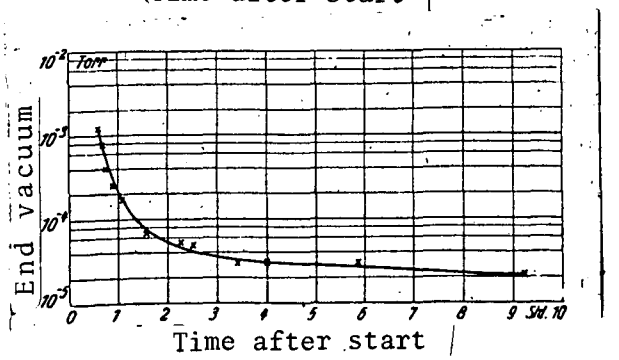

Figure 4. Single Phase Metal Oil Diffusion Pump (Leybold Model Q) Operating with Leybold Apiezonoil E. (Heating energy 320 W, end vacuum measured with an ionization manometer forevacuum pump Leybold Model XII).

TABLE IV. END VACUUM VALUES MEASURED IN A METAL DIFFUSION PUMP WITH AN IONIZATION MANOMETER

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Oil Filling	Apiezonoil B Leybold-Apiezonoil E	Diamyl phthalate*	Tricresyl Phosphate **	Octoil	Machine Oil (Shell CY2)
End Vacuum in Torr	$1.2 \cdot 10^{-5}$	$8 \cdot 10^{-5}$	1 to $2 \cdot 10^{-4}$	$4 \cdot 10^{-5}$	approx. 10^{-4}

Indicated as driving media for diffusion by:
* K. C. D. Hickmann, J. Frank Inst. 221 (1936), 383
** K. C. D. Hickmann, J. Phys. Chem. 34 (1930), 637.

3. Operating Properties of Driving Media Apiezonoil and Octoil

It has been established that in the most favorable situation with a specific driving medium the achievable end vacuum values depend upon the pump arrangement and the operating conditions (forevacuum and heat energy). Below, we will report as to how the end vacuum value may be influenced, or diminished by the conditions of the technical operation, air leaks, ssustained operation, aging of the oils and influence of water vapor.

a) Air Leaks

In the operation of a diffusion pump it is in many instances impossible to avoid cases in which the high vacuum-side of the pump, that is, the one coupled to the instrumentation, occasionally suffers one or more serious air leaks.

I. Severe Air Leaks

It may occur as a consequence of a poor seal in the connected instrumentation that the pressure suddenly increases to 10 Torr, 400 Torr or even to atmospheric pressure, and that the leak can be eliminated only after a few minutes or that the pump can be disconnected from the instrumentation only after several minutes. Often the opinion exists that as a result of such an air leak the oil pump will burn and consequently be completely destroyed. This does not happen at all, since the oil as a consequence of the described distillation process will self-clean in time inside the pump.

We have tried to determine the relationships of such air leaks by disconnecting the diffusion pump from the forepump, and allowing air to enter the hot pump at a given pressure (10, 420 or 760 Torr) and after several minutes (2 respectively 10) reconnecting the forepump. Figures 5 and 6 show the increase of the end vacuum after the reconnection of the forepump. After a maximum of one hour the pressure assumed in all instances the magnitude of 10^{-5} Torr once again, which for all practical purposes should be quite sufficient. Table V /184
reveals further which pressures were achieveable after longer pumping times. According to it, after air leakage at 10 or 420 Torr the same end vacuum result will be reached as before the leak. The situation with Apiezonoil in cases of such air leaks (10 to 420 Torr) is somewhat more favorable than with Leybold-Apiezonoil[10], while in the case of serious air leaks both Leybold-Apiezonoil and Apiezonoil B had the same result.

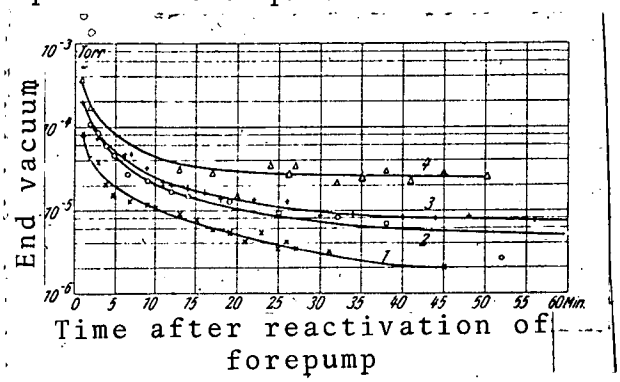

Figure 5. Recovery of the End Vacuum After Air Venting with Apiezonoil B in 2-stage Metal Diffusion Pump (Leybold Model P). (Heat energy 450 W, end vacuum measured with the molecular vacuum gauge, forevacuum pump Leybold Model V). Air Venting; 1 v 10 Torr 10 minutes; 2 o 420 Torr 2 minutes; 3 + 760 Torr 2 minutes; 4 Δ 760 Torr 10 minutes.

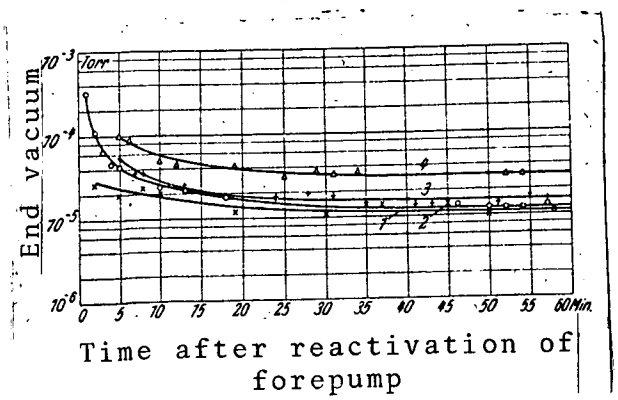

Figure 6. Recovery of the End Vacuum After Air Leak with Octoil and a 2-stage Metal Diffusion Pump (Leybold Model P). (Heat energy 250 W, end vacuum measured with the molecular vacuum gauge, forevacuum pump Leybold Model V). Air Leak: 1 x 10 Torr 10 minutes; 2 o 420 Torr 2 minutes; 3 + + 760 Torr 2 minutes; 4 Δ 760 Torr 10 minutes;

[10]These measurements were performed in 1941 with Leybold-Apiezonoil E. In the meantime the Leybold-Apiezonoil has been improved.

TABLE V. REQUIRED PUMPING TIME TO ACHIEVEMENT OF A DEFINITE END VACUUM VALUE AFTER AIR LEAKAGE IN METAL DIFFUSION PUMPS (Measured with the Molecular Vacuum Gauge)

Oil-Filling / Pressure and Duration of the Air Leak	An End Vacuum of x Torr was Reached in y Minutes After the Air Leak					
	Leybold-Apiezonoil E		Apiezonoil B		Octoil	
	x	y	x	y	x	y
10 Torr 10 min.	$5 \cdot 10^{-6}$	85	$2 \cdot 10^{-6}$	45	$1 \cdot 10^{-5}$	70
420 Torr 2 min.	$6.7 \cdot 10^{-6}$	240	$2.7 \cdot 10^{-6}$	120	$1.1 \cdot 10^{-5}$	120
760 Torr 2 min.	$7 \cdot 10^{-6}$	335	$1 \cdot 10^{-5}$ *	70	$1.2 \cdot 10^{-5}$	140
760 Torr 10 min.	$1 \cdot 10^{-5}$	240	$2.5 \cdot 10^{-5}$	50	$2.5 \cdot 10^{-5}$	100

*The end vacuum varies strongly after the air leak.

II. First Increase of the End Vacuum After A Serious Air Leak

For many purposes it is of less significance what pressure has been reached after long pumping times than how fast will the vacuum develop in the first minute after an air leak has taken place. The relationships shown under I are substantially for large high-vacuum instruments (distillation apparatus, high-voltage systems, mirror-coatings, etc.), where extremely long pumping times are used. The ratios during the first few minutes, however, are very important for rapidly occuring pumping processes as for instance in automatic pumping systems.

Figure 7 shows the increase of the vacuum in the first minutes after air leakage and the reconnection of the forepump. The measurements for Figure 7 were taken in the same way as described under I. A substantial advantage of a pump with separate distillation flasks in accordance with Figure 2 is demonstrated by the first increase of vacuum after air leakage. Figure 8 shows the first increase of vacuum for such a pump. As a comparison between Figure 7 and Figure 8 shows, the vacuum increases substantially quicker with a pump according to Figure 2. It should be also considered that in Figure 8 the pressures under 10^{-3} Torr were measured with the ionization manometer and the

Figure 7 pressures with the molecular vacuum gauge. According to Table I the measurements with the ionization manometer yield substantially larger pressure values under essentially similar circumstances.

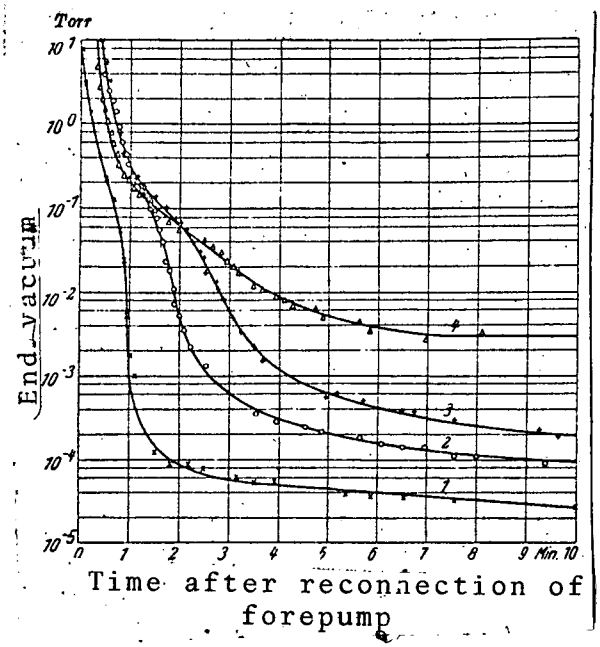

Figure 7. Initial Increase of the End Vacuum After Air Leakage with Leybold-Apiezonoil E in a Single-Stage Metal Diffusion Pump (Leybold Model Q). (Heat energy 320 W; end vacuum measured between 10 and 10^{-3} Torr with thermoelectrical vacuum guage, between 10^{-3} and 10^{-5} Torr with a molecular vacuum gauge; forevacuum pump/ Leybold Model V). Air venting: 1 x 10 Torr 2 minutes; 3 + 60 Torr 2 minutes; 4 Δ 760 Torr 10 minutes.

III. Gas Leaks (Small Air Leaks)

Finally we studied the problem of how oils behave, not when the pressure in the pump because of leaks might increase a few Torr, but what happens when for instance because of a gas leak in the installation for a short time (3 minutes) values of 10^{-1} Torr or 10^{-2} Torr are assumed. Figure 9 shows the recovery of the vacuum after such gas leaks. /185

b) Durability in Continuous Operation

While operating under higher pressure (for instance, 10^{-3} Torr on the suction side) substantial amounts of air are sucked in through the hot side of the pump. It may be imagined that under these conditions the oil would gradually oxidize after long continuous operation and consequently decrease the quality

of the end vacuum. For the verification of this question we filled a diffusion pump series of two metal pumps with Apiezonoil B, and introduced continuously through a needle valve a certain amount of air on the suction side so that the pressure at that point stabilized at approximately 10^{-3} Torr. This set-up was kept in operation for approximately 300 hours and we continued to measure the end vacuum of the enclosed needle valve at regular intervals. We were not able to determine in the 300 hours any measureable decrease of the quality of the end vacuum.

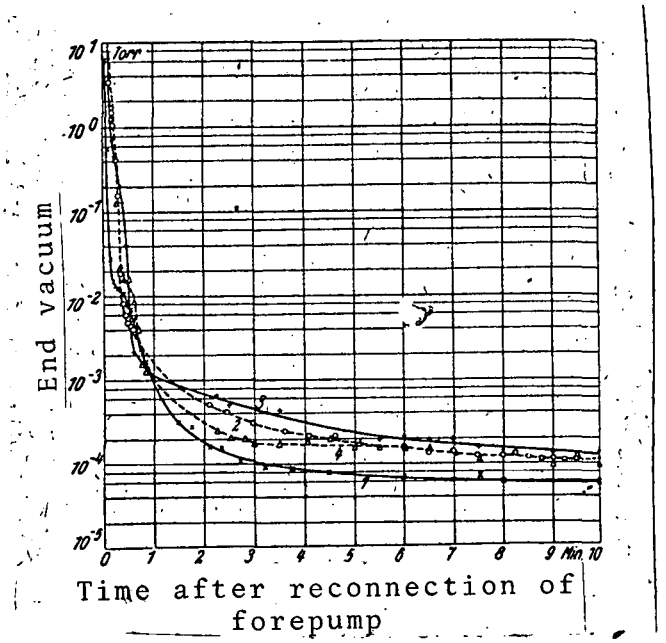

Figure 8. Initial Increase of the End Vacuum After Air Leakage with Leybold-Apiezonoil E in a 3-stage Metal Oil Diffusion Pump with Separate Distillation Flasks and Oil Reflux to the Second Stage (Leybold Model Q). (Heat energy 420 W; end vacuum measured between 10 and 10^{-3} Torr with a thermoelectric vacuum gauge, between 10^{-3} and 10^{-5} Torr with an Ionization manometer; forevacuum pump Leybold Model XII). Air venting: 1 v - — 10 Torr 10 minutes; 2 0--- 420 Torr 2 minutes; 3 + —— 760 Torr 2 minutes; 4 Δ--- 760 Torr 10 minutes.

c) Aging Stability

To determine whether a long exposure to atmospheric air would cause the oils to be attacked, we filled a metal pump with Leybold-Apiezonoil E, measured the end vacuum after cooled air was let into the pump and after a long period of time repeated the measurement of the end vacuum.

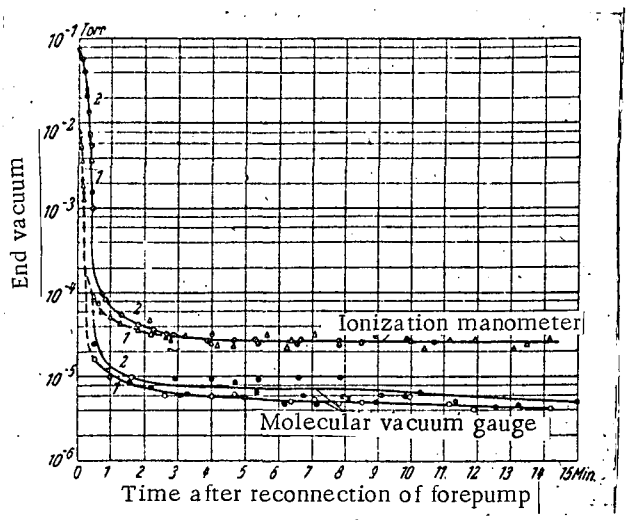

Figure 9. Recovery of the End Vacuum after a Gas Leak (Small Air Leak) in Leybold-Apiezonoil E and a Single-Stage Metal Diffusion Pump (Leybold Model Q). (Heat energy 320 W; end vacuum measured between 10^{-1} and 10^{-3} Torr with thermoelectric vacuum gauge, between 10^{-3} and 10^{-6} Torr with molecular vacuum gauge and ionization manometer; forvacuum pump Leybold Model XII). Gas leaks: 1. 10^{-2} Torr 3 min Δ measured with a th ermoelectric vacuum gauge and ionization manometer; 0 measured with a molecular vacuum gauge; 2. 10^{-1} Torr 3 min ⊙ measured with thermoelectric vacuum gauge and ionization manometer; ⊠ measured with molecular vacuum gauge.

This process was repeated several times. Within a period of 8 weeks we could not determine a noticeable decrease in the quality of the end vacuum. It should be mentioned at this point however that the oils are light-sensitive. We recommend therefore that they be stored in dark bottles.

d) Water Vapor Resistance

Since Octoil is an ester of phthalic acid with a higher alcohol, we could expect a saponification by incorporation of the H_2O molecule in the presence of water vapor, that is a decomposition into phthalic acid and alcohol. During the evacuation of the containers, because of the water film in the walls, there is always a certain amount of water vapor. This could presumeably lead to unpleasant decomposition of the Octoil. The question of whether such a decomposition may in fact occur in the practical operation of a diffusion pump has been verified in that we introduced a certain amount of saturated water vapor

through a needle valve on the suction side of the metal diffusion pump operated with Octoil. The needle valve was set in such a manner that the pressure on the suction side of the pump was always approximately 10^{-3} Torr. After 8 hours of operation the needle valve was closed off and the increase of the vacuum measured. Pumping times from 60 to 90 minutes were all that was necessary to return to the original end vacuum.

A similar experiment conducted with Apiezonoil B also recovered the original end vacuum in approximately 60 to 90 minutes after the closing off of the needle valve. Moreover, we conducted the following experiments with Apiezonoil B. A metal oil-diffusion pump was treated with an emulsion of 48.5 cm^3 Apiezonoil B and 1.5 cm^3 H_2O, and we measured the increase of the end vacuum after resumption of the operation. Figure 10 shows the curve of the end vacuum with time.

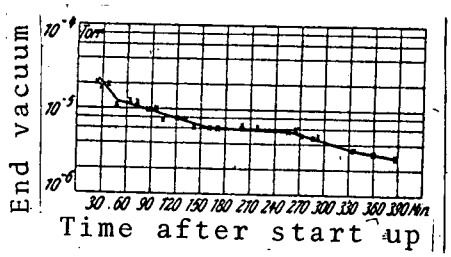

Figure 10. Two-stage Metal Oil Diffusion Pump (Leybold Model P) Operating with 48.5 cm3 of Water. (Heat energy 450 W; end vacuum measured with molecular vacuum gauge, forevacuum pump Leybold Model V).

After the tests with the metal oil-diffusion pumps it appears that both Apiezonoil B and Octiol seem to be absolutely water-vapor stable. As the following tests with glass pumps show, this applies to Apiezonoil as well. /186
In the case of Octoil the test in the glass pumps demonstrated that there is a substantial decomposition. The fact that this escaped observation in the tests with the metal pumps may mean that the Octoil used in the metal pumps had been exposed to a strong thermal decomposition (compare the end vacuum values in metal and glass pumps, Tables I and II). As a consequence the end vacuum deteriorates constantly and completely obscures the influence of the water vapor.

The tests with Octoil in the glass pumps were conducted in such a manner that a glass diffusion pump series was filled with Octoil and the end vacuum simultaneously measured with a molecular vacuum guage and an ionization manometer on the high vacuum side. After our experiences with the strong

decomposition of the Octoil in the operation of metal pumps we exercised extreme care. Before the heat-up of the diffusion pumps we removed the last traces of air dissolved in the Octoil by a thorough evacuation with the forepump. Only then did we connect the diffusion pumps and proceed with a gradual increase of the heating energy. Table VI, column 1 shows the end vacuum values obtained. A comparison with the last line of Table I reveals that in these carefully conducted experiments with Octoil and glass pumps during the ionization manometer measurement even smaller end vacuum values were obtained than with Apiezonoil, in contradiction to the results with the metal pumps. Then we introduced through a needle valve a certain amount of air on the high vacuum side of the glass diffusion pump series and regulated it to a pressure of some 10^{-3} Torr. After blowing air through the pump series we closed the needle valve and again measured the end vacuum contained. Table VI, column 2, shows that the same end vacuum was not reached as before (Table VI, column 1). A longer pumping improved the end vacuum value considerably (Table VI, column 3), but the original end vacuum values were not reached again. After these tests we found in the forevacuum tube of the glass diffusion pump long acicular crystals of phthalic acid anhydridc. The Octoil was apparently decomposed under the influence of the blown air and the moisture contained therein by splitting free phthalic acid.

TABLE VI. OIL DIFFUSION PUMP SERIES OF JENA GLASS OPERATED WITH OCTOIL

(Heat Energy 300 Watts, 150 Watt per Individual Pump, Forevacuum Pump Leybold, Model V).

End vacuum measured with:	Immediately following careful degasification and slow heat up, Torr.	After the blowing of air through the heated pump series, Torr.	After additional 30 hours of operation of the pump series to end vacuum, Torr
Molecular vacuum gauge	$2.1.10^{-6}$	$4 \cdot 10^{-6}$	$2 \cdot 10^{-6}$
Ionization manometer	$1.1 \cdot 10^{-6}$	$6.5.10^{-6}$	$2.2 \cdot 10^{-6}$

In contrast to this behavior of Octoil Apiezonoil B in glass pumps showed the same end vacuum value, as measured by the ionization manometer, of $1.7 \cdot 10^{-6}$ Torr after the blowing through of air. Apiezonoil is therefore not affected by moisture in the air.

Summary

The indication of the end vacuum value is for a particular driving medium is substantially dependent upon the method of vacuum measurement, so that the indications of different authors, employing in fact the same measurement methods (ionization manometer) but different types of tubes, do not offer a comparison. The physically most reliable values are given by measurement with an absolute manometer (molecular vacuum gauge). The value of the end vacuum is moreover dependent on the operating conditions (forevacuum and heating). The deciding influence on the level of the vacuum obtainable, moreover, is the type of pump installation (metal or glass diffusion pumps, single pumps or pump series). Since the oils are sensitive to oxidation and thermal decomposition, the highest possible end vacuum value will not be reached after an air leak. The distillation processes in the pump resulted in a self-cleaning of the oils so that after a certain pumping time we can again achieve the original end vacuum value. Relative to their insensitivity to thermal decomposition and oxidation we may consider the pure hydrocarbon oils (Apiezonoils) superior to the esters of organic acid (Octoil and Octoil S). They deliver a higher end vacuum in metal diffusion pumps and are moreover more resistant to water vapor influences. A type of oil diffusion pump is described wherein the distillation processes are scrupulously utilized for the cleaning of the oils. These pumps deliver higher end vacuum values and may be operated for modest requirements even with ordinary machine oil. After air leaks these pumps have an ability to recover the end vacuum quickly.

I wish to express my thanks for the tireless careful preparation of countless measurements to Miss E. Gunther and to Messrs. A. Heimsoeth and H. Schwarz.

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